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AEROSOL COMPOSITION
IN THE
MARINE AND COASTAL BOUNDARY LAYER*

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by
E.J. Mack, J.T. Hanley, R.J. Pilié, C.K. Akers and B.J. Wattle

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Aerosol composition in the marine and coastal boundary layer

E.J. Mack, J.T. Hanley, R.J. Pilie, C.K. Akers and B.J. Wattle

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Abstract

As part of a number of broader-based studies, Calspan acquired a considerable quantity of data describing boundary-layer aerosol characteristics both at sea and in maritime continental locations. This paper focuses on aerosol composition data obtained in the North Atlantic, in the Mediterranean, in the northern Gulf of Mexico, off the coasts of California, Nova Scotia and Portugal, and on shore on Cape Cod and on the North Sea Coast of West Germany. The data show that the marine aerosol population varies considerably in composition, particularly in coastal areas, and does not necessarily comprise primarily sea salt aerosols. A continental/anthropogenically-derived component to the marine aerosol population is generally always observed, even in remote marine areas. In coastal regions, dramatic changes in aerosol composition occur as a result of wind shifts or air mass changes. As a result of these compositional differences, response of the aerosol to fluctuations in relative humidity is expected to differ from one locale or air mass to another. Therefore, aerosol size spectra alone are not sufficient for the prediction of visibility or the potential performance of EO systems under changing humidity conditions.

Introduction

During nine field studies conducted since 1974, a considerable quantity of data was acquired describing the aerosol characteristics of the marine boundary layer in the coastal areas of Cape Cod and West Germany, off the west coasts of the United States and Europe, off the coasts of New England and Nova Scotia, in the northern Gulf of Mexico, in the Mediterranean and in the mid-Atlantic. These field studies included participation in Panama City I and II, CEWCOM-76 and -78, and Meppen-80. Observations of aerosol size spectra (0.01-30.0 μm diameter), cloud condensation nuclei, bulk aerosol composition, individual particle ($>0.2 \mu\text{m}$ diameter) composition, visibility and relative humidity and other meteorological parameters were acquired in the lowest 20 m of the marine atmosphere. The data show that the marine aerosol population varies considerably in number concentration, size spectra and composition, particularly in coastal areas, and does not necessarily comprise primarily sea salt aerosols. The data demonstrate that what is thought to be "clean" (natural) marine aerosol at one location should not be extrapolated to describe other "clean" marine situations elsewhere nor necessarily be termed "natural".

Instrumentation utilized in these studies included: a TSI Electrical Aerosol Analyzer and Calspan impactors for aerosol and sea spray size spectra (0.01-30 μm diameter), a Gardner Small Particle Detector for total aerosol concentration, a Calspan thermal diffusion chamber (static) for cloud condensation nucleus (CCN) activity spectra, an MRI Integrating Nephelometer for visibility and scattering coefficient, Battelle-type and Casella cascade impactors to collect samples of aerosols for individual particle analysis via scanning electron microscopy (SEM) and elemental energy dispersive x-ray analysis (EDXA), 10-vol filter samples (Fluoropore teflon-membrane, 0.5 μm pore size) for mass-loading and bulk aerosol chemistry via wet-chemical and XRF analysis, and standard meteorological instrumentation for winds, temperature, dewpoint, etc. Detailed descriptions of specific cruise objectives, cruise scenarios, instrumentation set-up, analysis procedures, and data sets may be found in References 1-11. The purpose of this paper is to provide a summary of these data.

Aerosol concentrations

Typical and average values for aerosol and CCN concentrations observed (hourly measurements obtained in the absence of fog and storm conditions) during the indicated studies are summarized in Table 1. [The Gulf of Mexico data are averaged for on-shore (marine) and off-shore (continental) wind conditions, while other data show the general range of temporal variations in aerosol concentrations.] Shown in Table 1 are number concentration values for total (Aitken) particulates, for aerosols greater than 0.1 and 1.0 μm diameter, and for CCN active at 0.2% and 1.0% supersaturation. These data show that, in the coastal areas (within 300 km offshore but away from the immediate coastline), total particle concentration is typically $<6000 \text{ cm}^{-3}$, that ~25-50% of the particles are in the size range 0.1 to 1.0 μm diameter, with the remainder being $<0.1 \mu\text{m}$ in diameter, and that ~50% of the particles are active at 1.0% supersaturation. Maximum particle concentrations, and greater fluctuations in CCN, RH and visibility are found in immediate coastal areas. Total-particle

Table 1. Observed Aerosol Concentrations in the Marine Boundary Layer*

		Visibility (km)	RH (%)	Total Particle Conc. (cm^{-3})	Particle Concentration at Diameters		-----CCN @-----	
					>0.1 μm (cm^{-3})	>1.0 μm (cm^{-3})	0.2% SS (cm^{-3})	1.0% SS (cm^{-3})
Coast of Portugal (within 1200 km offshore)	May 77	20-50	75-90	900-5000	400-2000	0.9-3.0	300-500	600-1000
Mediterranean (150-250 km offshore)	Jun 77	25-60	65-80	800-3500	250-1500	0.7-3.0	150-900	400-2800
Coast of Nova Scotia (within 150 km offshore)	Aug 75	-	-	400-2000	-	-	130	450
New England Coast (200-400 km offshore)	Aug 75	-	-	2000-6000	-	-	580	1350
New England Coast (within 300 km offshore)	May 77	20-80	65-75	4000-15000	1000-5000	0.3-6.0	350	990
Gulf of Mexico (20 km offshore)								
Marine air (Panama City, FL)	Feb 77	43	65-95	3400	-	0.1-1.3	-	-
Marine air	Nov 78	23	75-95	1700	300-1200	1.0-10.0	730	1450
Continental air	Feb 77	32	40-75	5500	-	0.1-1.2	-	-
Continental air	Nov 78	20	35-85	3800	1500-3500	0.1-1.0	1010	2130
Offshore Coast of S. California (within 150 km)	Oct 76	10-40	70-95	400-4000	-	-	50-1000	400-2200
(within 150 km)	Nov 78	10-30	75-95	300-10000	100-2000	6.0	100-700	300-3500
(beyond 150 km)	Sep 76	30-80	70-95	<200-600	-	-	20-200	100-400
Off N. California (100-150 km)	Aug 74	-	-	-	-	-	60	200
Mid-Atlantic	May 77	80	50-75	<200-500	30-150	0.8-4.0	60-140	90-200
Cape Cod (12 km inland)	Jul 80	4-60	50-95	4000-35000	500-4000	0.5-100	60-600	400-2000

*Single numbers represent averages; multiple figures are typical range values and do not include extremes.

and CCN concentrations generally decrease with distance offshore, although high concentrations are often observed well at sea as was the case off the coast of Portugal. Minimum particle concentrations observed in the mid-atlantic and far-offshore West Coast may be representative of "clean" marine conditions. At any particular location, however, observed aerosol concentrations exhibit temporal variations of nearly an order of magnitude.

The chemistry of aerosols in the marine atmosphere

In the boundary-layer atmosphere, particularly in coastal areas, the composition (as well as the size spectra) of ambient aerosols is dependent on air mass history and wind trajectories. The influence of the land/sea breeze winds on the composition of aerosols observed in coastal areas is illustrated by the data presented in Figure 1. In the figure, the airborne concentrations of selected constituents of aerosols sampled several times each day during a 4-week period at an offshore platform located in the Gulf of Mexico 20 km southwest of Panama City (Florida) are plotted as functions of wind direction. The samples were collected on 47 mm Teflon membrane filters (0.5 μm pore size) with flow rates of ~ 30 l/min. The results of XRF analyses of the filter samples were averaged for 45° increments in wind direction. (At the platform, winds from the southerly direction between 150° and 260°T have substantial marine fetch, while northerly winds from 315°T to 090°T have a maximum of 40 km fetch over water after leaving the continent.) The data show that with the sea breeze and increasingly maritime air (i.e., with more southerly winds) in the afternoons, airborne concentrations of Cl, Na, and Mg (the major constituents of seawater) increased by about an order of magnitude, while concentrations of Si and Al (both originating in soils) decreased. Conversely, more continental aerosols were observed in the coastal region under land breeze conditions. (Sulfur is the fourth most abundant constituent of seawater, but it is also a major component of continental aerosols and hence did not on the average vary significantly in the coastal zone.)

Temporal variations in aerosol composition also occur as a result of air mass changes as is illustrated in Figure 2. The figure presents the results of SEM and EDXA analyses of individual particles collected via cascade impactor at Meppen, FRG (located ~ 80 km inland from the North Sea). Note the changes in aerosol composition-frequency which occurred as a result of major air mass changes of 7 and 11 November and of wind shifts on 8, 10 and 12 November. During the period 7-11 November, we experienced an aged continental air mass which was trapped between two east-west frontal zones over central Europe and whose aerosol burden was dominated by ammonium sulfate. For the four days prior to 8 November, steady northeasterly winds (produced by high pressure over Scandinavia) brought in air dominated by flyash originating in the industrial regions of northern Europe. After 11 November, a succession of frontal passes brought in air masses from the North Atlantic which experienced different residence times over western Europe or the British Isles and whose aerosol burdens included mixtures of sea salts (NaCl) and soil silicates (not flyash). The dramatic increase in NaCl aerosols to nearly 100% of the population of 12 November was correlated precisely with a frontal passage and a brief wind shift to northwesterly which brought marine air (with the shortest possible over-land fetch) directly over the field site.

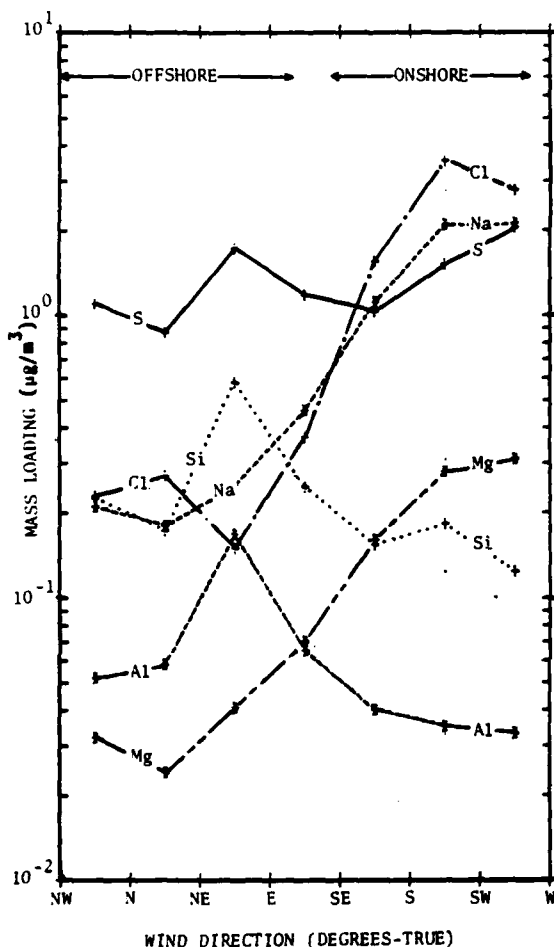


Figure 1. Average Airborne Concentration of the Elemental Constituents of Aerosols Observed 20 km Offshore in the Northern Gulf of Mexico, November-December 1978

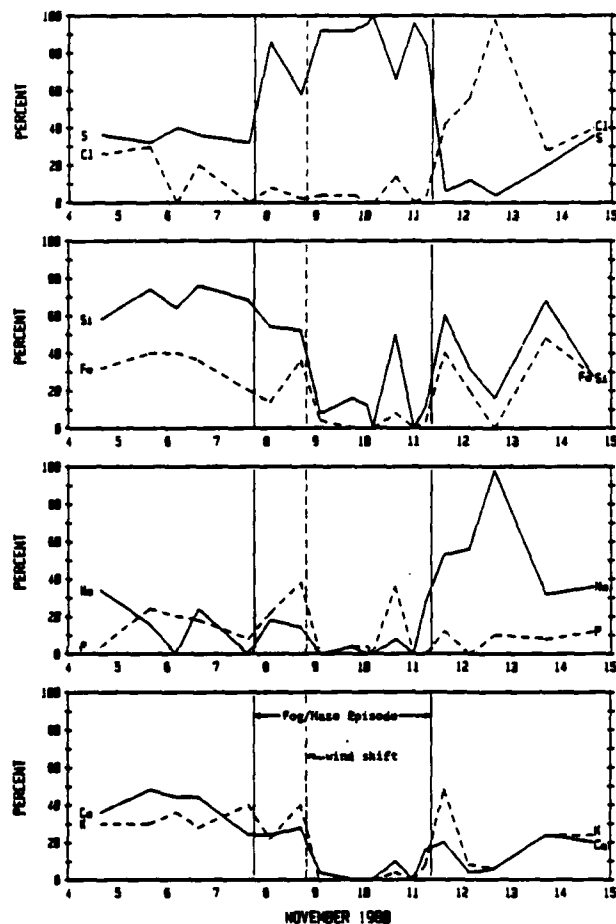


Figure 2. The Percentage of Sampled Aerosols (0.3-10 μ m diameter) Containing Selected Chemical Elements as a Function of Time at Meppen, West Germany 4-14 November 1980.

Observed mass loading (airborne concentration) data are summarized for different maritime locations in Table 2. It is readily seen that average concentrations of all constituents of the observed aerosols differed by approximately a factor of 10 between locations. Note that measurable concentrations of such continental materials as Al and Si are found far at sea while concentrations of Cl can be quite small in the marine environment. Note that, comparing even the previously defined "clean" marine locations (mid-atlantic and far offshore California), mass loading of the various constituents varies considerably.

The ratio of the concentration of elemental constituents to that of sodium in ambient aerosols in the marine boundary layer, when compared to that of sea water, provides a useful indicator of the continental or maritime characteristics of the aerosol. For example, the sodium ratios for Al and Si in bulk sea water are extremely small (i.e., $\sim 10^{-4}$), and any detectable airborne quantities in the marine atmosphere must be attributed to continental sources. Further, the sodium ratios for Cl and Mg in sea water are relatively constant, and aerosol values which approach those of sea water are indicative of sea salt aerosols. Sodium ratios for the data presented in Table 2 are compared against those of sea water to produce enrichment ratios (factors) which are tabulated in Table 3. The enrichment ratio

Table 2. Average Airborne Concentrations of Selected Constituents of Marine Boundary Layer Aerosols

			Absolute Concentrations ($\mu\text{g}/\text{m}^3$)										
			Na	Cl	S	SO ₄ --	Mg	K	Ca	Al	Si	Mn	Fe
Off N. American Coast (Atlantic)	May 77	(within 300 km)	1.0	2.81	1.4	1.4	.38	.08	.44	.22	.44	.01	.50
New England Coast	Aug 75	(200-400 km offshore)	1.3	.02	-	14.0	.12	.22	.13	.26	-	-	-
Off Portuguese Coast	May 77	(within 1200 km)	0.2	0.17	1.5	2.1	.08	.03	.09	.05	.11	.02	.06
Mediterranean	Jun 77		0.2	0.17	1.7	3.9	.09	.04	.14	.07	.15	.02	.10
Off Nova Scotia	Aug 75	(within 150 km)	0.9	< .02	-	4.0	.05	.19	.09	.22	-	-	-
N. Gulf of Mexico (20 km offshore)													
on-shore wind	Nov 78		1.8	3.01	1.4	-	.25	.12	.17	.03	.16	-	0
off-shore wind	Nov 78		0.2	0.27	1.1	-	.04	.06	.07	.09	.29	-	.06
Coast of South California													
within 100 km	Oct 76		2.1	1.20	-	8.4	.31	.20	.19	.013	-	-	-
within 150 km	May 78		2.1	4.35	0.4	-	.35	.36	.62	.10	.54	0	0
beyond 100 km	Sep 76		2.4	3.10	-	2.5	.30	.14	.08	.004	-	-	-
Mid-Atlantic	May 77		0.4	1.38	0.3	0.3	.10	.02	.06	.02	.03	0	.02

Table 3. Enrichment Ratios (Relative to Sodium Ratios of Sea Water) for Selected Constituents of Marine Boundary Layer Aerosols

			Enrichment Ratios $\frac{X/\text{Na (Sample)}}{X/\text{Na (Sea Water)}}$									
			Cl	S	SO ₄ ⁻⁻	Mg	K	Ca	Al ($\times 10^5$)	Si ($\times 10^3$)	Mn ($\times 10^5$)	Fe ($\times 10^4$)
Off N. American Coast (Atl)	May 77	(within 300 km)	1.6	17.0	5.6	3.2	2.0	11.0	2.2	1.5	0.5	0.5
New England Coast	Aug 75	(200-400 km offshore)	< .01	-	43.0	0.8	4.3	2.5	2.0	-	-	-
Off Portuguese Coast	May 77	(within 1200 km)	0.5	89.0	42.0	3.3	3.8	11.0	2.5	1.8	2.5	2.5
Mediterranean	Jun 77		0.5	101.0	78.0	3.8	5.0	18.0	3.5	2.5	2.5	2.5
Off Nova Scotia	Aug 75	(within 150 km)	< .01	-	18.0	.5	5.3	2.5	2.4	-	-	-
N. Gulf of Mexico (20 km offshore)	Nov 78	on-shore wind	0.9	9.5	-	1.2	1.8	2.3	0.2	0.3	0	0
	Nov 78	off-shore wind	0.6	55.0	-	1.3	6.3	7.8	3.6	4.1	0	0.3
Coast of South California												
	Oct 76	within 100 km	0.3	-	16.0	1.3	2.6	2.4	.06	-	-	-
	May 78	within 150 km	1.2	2.2	-	1.4	4.7	7.9	0.5	0.9	0	0
	Sep 76	beyond 100 km	0.7	-	4.2	1.1	1.6	0.8	.02	-	-	-
Mid-Atlantic	May 77		1.9	9.5	3.2	2.1	1.3	3.8	0.5	0.3	1.5	1.3

(E) is simply the sodium ratio of an element (X) in a sample divided by the sodium ratio for that species in sea water. If the sample is pure marine (sea salt) aerosol, the

$$E(x) = \frac{(x/\text{Na})_{\text{sample}}}{(x/\text{Na})_{\text{sea water}}}$$

enrichment ratio is 1. (Due to limitations and inaccuracies imposed by sampling conditions, sample handling, trace ambient concentrations, filter background and analysis procedures, values ranging from ~0.7 to ~1.5 may be indicative of dominant marine.) Enrichment ratios >1.5 indicate an excess of specific species (relative to sodium) from either fractionation processes at the sea surface or, more likely, continental sources.

All elements considered, the analysis presented in Table 3 suggests that, averaged over the sampling times and respective cruise areas, a pure marine aerosol burden (i.e., exclusively sea salt) was not observed at any time during the at-sea studies. The closest approach to pure marine aerosol was observed off the coast of southern California (beyond 100 km) in September 1976; but even there a substantial excess of sulfate aerosol matter was observed. The data show that even over the open remote ocean, there are always at least background levels of materials of continental/anthropogenic origin (e.g., S, Al, Si, etc., including heavy metal catalysts such as Fe and Mn) in the bulk aerosol observed in the marine atmosphere and that relative concentrations can vary over a considerable range.

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We have also collected aerosol samples with 6-stage Battelle and 4-stage Casella cascade impactors for analysis via SEM and EDXA. The combination of these two techniques allows (1) visualization of the impacted particle where size measurements can be obtained and (2) determination of elemental composition of individual particles with specific identification of elements from sodium (atomic number 11) and greater in atomic number. (Data can be obtained only for particles $>0.2 \mu\text{m}$ diameter; maximum particle sizes are generally $<15.0 \mu\text{m}$.) The samples are collected on cellulose acetate propionate substrates with flow rates of 12 l/min and sampling times of $\sim 2-30$ minutes.

From SEM analysis, as in Figure 2, it was found that individual particles could be grouped into five composition categories according to total elemental composition:

- (1) those with atomic numbers lower than Na--probably organics;
- (2) NaCl only--sea salt;
- (3) NaCl mixed with other inorganic salts;
- (4) inorganic salts without NaCl--sulfates, calcite, gypsum, etc.;
- (5) and Si containing compounds--flyash and soil particles.

The percentages of particles found in each of these five composition groups at different locales are summarized in Table 4. Inspection of the data again reveals that aerosol populations differ dramatically in composition, comparing one location to another. At times in the marine atmosphere, "pure" NaCl aerosols can account for 80% of the aerosol population ($>0.2 \mu\text{m}$ diameter); at other times only 25% of the particles are NaCl; and in coastal regions as few as 10% of the aerosols may be NaCl. In the absence of high proportions of NaCl particles, aerosols composed of elements with atomic numbers lower than Na (probably organics) can comprise 15-35% of the ambient aerosols. Likewise, other inorganic salts, in particular ammonium sulfate and probably nitrates, can account for 15-45% of the aerosol population, particularly in coastal areas. A background level of silicate-containing aerosols (mixtures of flyash and soil silicates accounting for 2-20% of the aerosols) is always found; in coastal areas, silicates can comprise nearly 50% of the aerosol burden.

Differences in the composition of ambient aerosol burden, such as documented above, suggest that the response of the aerosol population to changes in relative humidity will differ from time to time and from one locale to another in a manner not predictable from aerosol size spectra data. Such aerosol growth differences are illustrated in mass loading data obtained at Meppen, FRG and presented in Figure 3. During the Meppen field study, aerosol samples were quantitatively collected on 37 mm Fluoropore PTFE (polytetrafluoroethylene) filters, $0.5 \mu\text{m}$ pore size. Filters were pre-weighed at 35% RH, pre-mounted and sealed in individual holders, and exposed during evening hours at Meppen. Upon return to the laboratory, the filters were again weighed at 35% RH to determine the dry weight of the aerosol collections and, hence, estimates of ambient "dry" mass-loading. Care was taken to insure that the samples had reached equilibrium at 35% RH before weighing.

The above described weight measurements were made with a Cahn Electrobalance Model 26. The sample filters were placed in an aluminum foil pan which is glued to a "stirrup-and-wire" rig suspended from the Cahn balance into a small chamber in which humidity can be controlled and monitored. This set-up can be used to obtain estimates of the change in mass loading of an aerosol sample as a function of relative humidity. Very briefly, humidity over the samples is lowered to $<5\%$ and then raised in steps to a maximum of $\sim 95\%$ RH; RH is then lowered step-wise. At each change in RH, the sample weight is monitored to insure that the sample has achieved equilibrium with ambient RH; a period of 5-15 minutes is usually required to achieve equilibrium, as indicated by no further change in weight of the sample. (The changes in weight due to hygroscopicity of some filter media and glues used in the filters and aluminum pan attachment are accounted for in the procedure.)

Six samples from the period at Meppen represented by Figure 2 were tested, using the above-described procedures, to produce the "growth" curves presented in Figure 3. For each of these analyses, RH was increased from $<5\%$ to 95% and then decreased to the RH at which complete drying occurred, all in steps of $\sim 10\%$ RH. Note that all of the curves reflect the well-known hysteresis effect. The deliquescence points of these samples are not precisely known because of the large RH-steps imposed by funding limitations; hence, the dashed portions of the curves for increasing RH connect the highest RH step achieved before deliquescence occurred and the lowest RH step achieved after deliquescence; the actual data points are shown in the figure. However, for these highly-mixed "natural" aerosol samples, deliquescence would be expected to occur over a broad range of RH, given the sulfate chloride and potential nitrate compounds observed in our samples.

Table 4. The Percentage of Particles in the Size Range 0.2-10.0 μ m Diameter as Functions of Composition and Sampling Location

		Organics (%)	NaCl (%)	Mixed: NaCl and Other Salts (%)	Other Salts Without NaCl (%)	Si (%)	Number of Particles Counted
Coast of Portugal (within 1200 km offshore)	May 77	33	25	17	13	12	200
Mid-Mediterranean	Jun 77	17	22	8	44	9	400
New England Coast (within 300 km offshore)	May 77	2	81	11	3	2	150
Gulf of Mexico							
Marine air (20 km offshore)	Nov 78	14	80	1	2	3	240
Continental air (20 km offshore)	Nov 78	27	25	3	34	11	280
Coast of South California (within 150 km offshore)	May 78	39	15	1	24	21	1350
Mid-Atlantic	May 77	12	68	10	8	2	300
Otis AFB (Cape Cod)	Jul 80	21	9	8	16	46	360
Meppen, West Germany	Nov 80	1	10	6	47	36	675

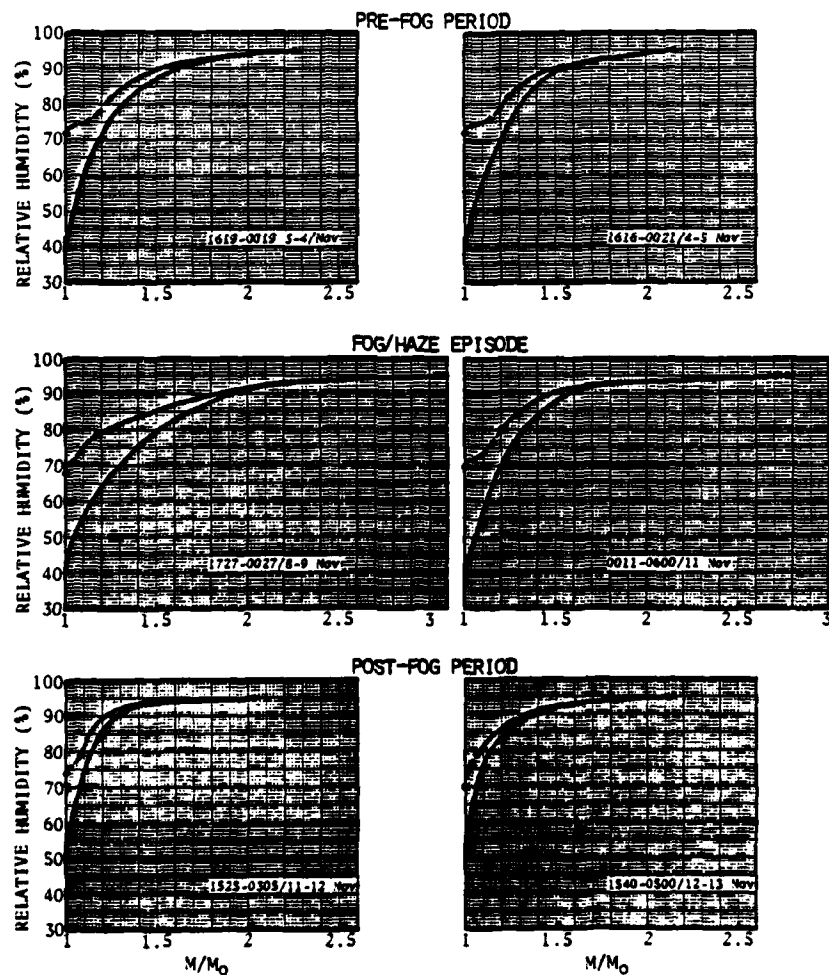


Figure 3. Relative Increase in Mass Loading (M/M_0) as a Function of Relative Humidity for Six Aerosol Samples Obtained at Meppen, West Germany, 3-13 November 1980.

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Note in Figure 3 that the growth curves of the two samples for each of the three previously defined meteorological periods exhibit similarities in the slopes of both growth and hysteresis, in maximum growth at 95% RH and in the RH required to completely dry the sample, implying some consistency within airmasses. However, inspection of growth curves for the three periods shows that the predominantly-sulfate aerosol population of the 7-11 November airmass, as a whole, apparently was capable of greater growth (and, hence, greater visibility restriction) with increasing relative humidity.

Conclusions

It must be recognized that the conclusions presented here are based on aerosol data acquired under specific meteorological circumstances (in the absence of fog and storm conditions) and what was observed is not necessarily typical in an absolute sense. However, the data demonstrate that the aerosol population in maritime locales can vary considerably in both size spectra and chemical composition, particularly in coastal areas and does not necessarily comprise primarily sea salt aerosols.

1. In the coastal marine atmosphere, total particle concentrations typically average $<6000 \text{ cm}^{-3}$. On the open ocean, total particle concentrations of $<500 \text{ cm}^{-3}$ are typical. Average concentrations of aerosols of $>1.0 \mu\text{m}$ diameter are usually $\sim 1 \text{ cm}^{-3}$ everywhere in the marine atmosphere. However, concentrations of aerosols at all sizes exhibit temporal and spatial fluctuations of an order of magnitude.

2. The marine aerosol, on the average, is not solely composed of sea salt. At times, sea salt aerosols can comprise 80% of the marine aerosol burden; at other times as little as 10%. There appears to always be a continental-source component to the marine aerosol burden. The continentally-derived materials most frequently observed in the marine atmosphere are of organic, sulfate, flyash, and common mineral origin (e.g., silicates, calcite, gypsum, etc.). As with number concentration, chemical concentrations (mass loading) exhibit temporal and spatial fluctuations of an order of magnitude.

3. Because marine aerosol composition, particularly of the larger more optically effective particles, varies considerably and is not necessarily sodium chloride, aerosol response to changes in relative humidity will vary from time to time and from one locale to another and will be other than that predicted from assumptions of sodium chloride composition. The primary conclusion, therefore, is that knowledge of the marine aerosol size spectrum alone is not sufficient to predict the behavior of electro-optical devices nor the response of those aerosols to changes in relative humidity.

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